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13. ABSTRACT *(Maximum 200 words)*

The goal of the project is to implement a novel type of optical detection system for simultaneous time-, frequency-, and space-resolved measurements of nanoscale phenomena: the Balanced-Homodyne Chronocyclic Spectrometer. This system uses balanced, optical homodyne detection, with a wavelength-tunable, pulsed local oscillator field, coupled with pinhole aperture spatial selection, to time resolve the spectrum of light emitted from photo- excited nanoscale structures. The main task for the grant period was the construction of the laser source and spectral filters for the chronocyclic spectrometer. This system consists of a self modelocked cavity dumped titanium:sapphire (TiS) laser which produces 50 nJ pulses of 13 fsec duration with a 100 nm bandwidth centered at 800 nm. This bandwidth is utilized to generate independently tunable pump and local oscillator pulses by splitting the oscillator output and spectrally filtering each of the two resulting beams. Construction of a TiS regenerative amplifier for situations where higher pulse energies are required is nearly complete. We have also developed pinhole mask fabrication techniques necessary to define a nanometer-scale region of optical excitation or signal collection in our samples.

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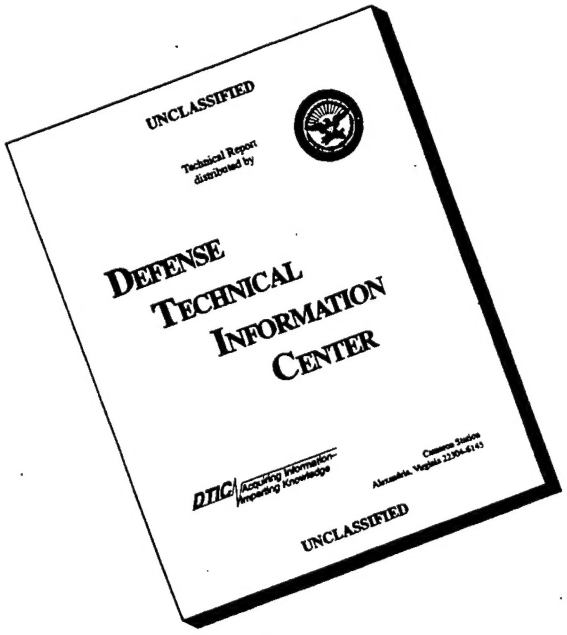

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A CHRONOCYCLIC SPECTROMETER FOR NANOSCALE STUDIES

FINAL PROGRESS REPORT

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STATEMENT OF THE PROBLEM STUDIED

The goal of the project is to implement a novel type of optical detection system for simultaneous time-, frequency-, and space-resolved measurements of nanoscale phenomena: the Balanced-Homodyne Chronocyclic Spectrometer. This system uses balanced, optical homodyne detection, with a wavelength-tunable, pulsed local oscillator field, coupled with pinhole aperture spatial selection, to time resolve the spectrum of light emitted from photo- excited nanoscale structures. The main task for the grant period was the construction of the laser source and spectral filters for the chronocyclic spectrometer. This system consists of a self modelocked cavity dumped titanium:sapphire (TiS) laser designed to produce 50 nJ pulses of 13 fsec duration with a 100 nm bandwidth centered at 800 nm. This bandwidth is utilized to generate independently tunable pump and local oscillator pulses by splitting the oscillator output and spectrally filtering each of the two resulting beams. To experimentally demonstrate the principle of the balanced homodyne chronocyclic spectrometer, a measurement example is described in which phase-randomized optical homodyne detection was used to characterize the time-frequency profile of a linearly chirped ultrashort laser pulse. The second task during the grant period was to develop the pinhole mask fabrication techniques necessary to define a nanometer-scale region of optical excitation or signal collection in our samples. A technique for producing the necessary masks utilizing liftoff processing with monodisperse polystyrene spheres has been developed. Finally, we have designed a TiS regenerative amplifier for situations where higher pulse energies are required. Construction of this system is nearly complete.

SCIENTIFIC PROGRESS AND ACCOMPLISHMENTS:

INTRODUCTION:

Light pulses from lasers or from laser-excited materials often contain temporal and spectral information on sub-ps time scales. Conventional methods for time resolving light intensity and spectra on these times scales include nonlinear optical techniques such as frequency-resolved optical gating (FROG)[1] and frequency upconversion in a nonlinear optical crystal.[2] These methods detect light in many spatial modes (the acceptance angle is large), and are therefore useful for detecting the average properties of a total signal field. In some cases, however, spatial mode selection may be desired. This could be achieved using an optical fiber as a spatial filter before the upconversion. A disadvantage of the nonlinear techniques is that they have low quantum efficiency, due to the nonlinear interaction required, meaning that not every signal photon is detected. This makes them impractical as a method for measuring the photon statistics of the signal in addition to its mean photon number and spectrum.

Photon statistics measurements are of interest for quantum-optical devices such as pulsed squeezed-light generators, for example parametric amplifiers.[3] Also of interest are sub-ps photon-statistics measurements of light emitted by semiconductor devices such as vertical-cavity surface emitting lasers (VCSEL).[4] In addition to photon-number statistics, the behavior of the optical phase of a signal field is of interest– for example, to determine whether a signal is phase coherent with the laser that produced it. The Raymer laboratory has recently pioneered the development of a technique, based on balanced-homodyne detection, for measuring photon statistics and optical phase statistics on such ultrafast time scales, and has applied the methods to studies of pulsed squeezing,[5] semiconductor lasers,[6] quantum wells,[7] and light scattering in random dielectric media.[8]

The chronocyclic spectrometer, built under the present grant, encompasses several research techniques already presented, namely Optical Homodyne Tomography (OHT) and Photon Number Sampling. The general layout for an ultrafast balanced-homodyne chronocyclic spectrometer is shown in Figure 1. There are two branches in our layout: the LO branch and the signal branch. The starting point for both is an ultrafast laser source. From this source we develop an LO field that has been spectrally filtered, time delayed, and phase adjusted. These elements can be programmed via computer and run synchronously with the laser source. The signal arm constitutes a light source that might be optically synchronized with the ultrafast laser source, electrically synchronized with it, or not synchronized at all (hence the dashed line). Only in the case of optical synchronization (such as squeezing) will the phase adjustment between the LO and the signal field be used (hence the dashed box). Those measurements will be phase-coherent with the LO and may be used to reconstruct fully the quantum state of the signal field (OHT). The signal fields that are not phase-coherent with the LO will yield measurements that are phase-random and may be used to reconstruct the photon number statistics. In this case, the phase adjustment is irrelevant.

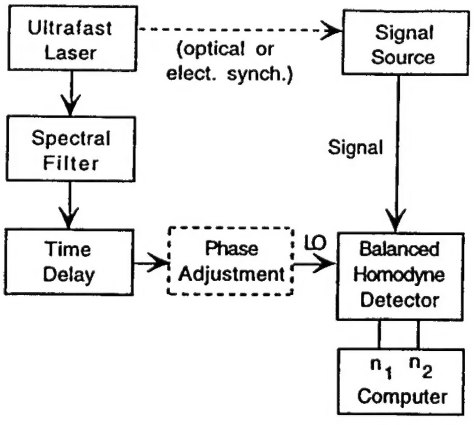

Figure 1: Elements of the Chronocyclic Spectrometer

The time resolution of the method can be sub-100 fs and the spectral resolution is Fourier-

transform limited. The method is linear in signal field strength, and the sensitivity is shot-noise limited, allowing less than one signal photon on average to be detected in a given time window. Finally, the method offers the powerful capability to determine not only the average photon number of a signal in a time-frequency window, but also its complete photon statistics.

As an alternative to standard photon-counting methods, balanced-homodyne detection offers higher quantum efficiency (QE), much faster sampling times (as short as the LO pulse), and spatial-mode selectivity. The technique can have quantum efficiency exceeding 99% (we typically operate at 85%). Even though the detectors used have a noise level of 200 electrons rms, the reconstructed state allows determination of photon distribution with good resolution between and in the range n=1-100. If one is only interested in mean signal levels, however (and not trying to distinguish between n and n+1 photons), the dynamic range is excellent, linearly detecting signals over five orders of magnitude, from 10^4 photons down to 0.1 photons.

A MEASUREMENT EXAMPLE:

With a prototype system (not having independent wavelength tunability of signal and LO beams) we have demonstrated the principle of this technique by measuring the complete time-frequency profile of a linearly chirped ultrashort pulse with a time resolution of 1 ps and a wavelength resolution of 1.2 nm.

The laser system consists of a Ti:Sapphire oscillator seeding a chirped pulse amplifier, using linear dispersion grating pairs to stretch the seed pulse before the amplifier and recompress the optical pulse after amplification. The pulse is spectrally filtered in the grating compressor with a narrow slit to give a spectral resolution of 1nm. The corresponding temporal width increases to 1.2ps, giving a pulse that is 40% above the transform limit. The slit is mounted on a translator, providing a tunable range from 820-840nm. The pulse then passes through a trombone (roof mirror on a translator) to give it an appropriate time delay. Finally, the pulse reflects from a mirror mounted on a piezoelectric translator (PZT) to give phase control. For this experiment we have employed a phase-random procedure, achieved by driving the PZT with a sine wave. At this point, the optical pulse constitutes our LO: spectrally filtered, temporally shifted, phase random. The LO and the signal are mixed on a 50/50 beamsplitter, detected and stored in the computer. A program analyzes the distributions and calculates the mean photon number in the signal.

In order to characterize the system, we used a known pulse as our signal. We inserted a beam splitter before the compressor pair to pick off a portion of the stretched pulse coming from the regenerative amplifier. This pulse is heavily chirped, spectrally broad and long in temporal duration. We first verified that the translation of the slit in the compressor led to a linear translation in the spectral window. We set the slit manually to 820nm and let the computer do a temporal scan (by moving the trombone) in 1ps steps. We then incremented the spectral window by 1nm and repeated the scan. We did this for 20 positions of the spectrum, from 820-840 nm, and 200 positions of time. The results are shown in Figure 2. The results show a pulse with linear chirp, ≈100 ps in duration, ≈10 nm in spectral width, consistent with our expectations.

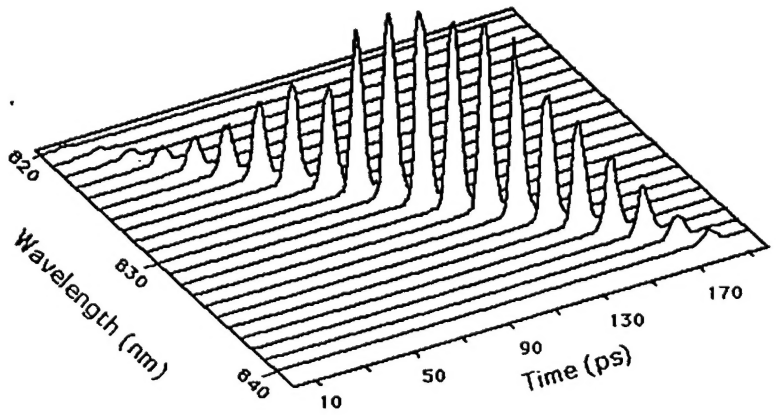

Figure 2: Time-wavelength profile of a linearly chirped, stretched laser pulse.

DESCRIPTION OF CAVITY-DUMPED Ti:S LASER AND SPECTRAL FILTERS

To realize the full potential of the balanced homodyne chronocyclic spectrometer, we have constructed a modelocked cavity dumped titanium:sapphire (TiS) oscillator and two spectral filters for generating independently tunable pump and local oscillator pulses. The modelocked cavity dumped oscillator produces pulses of 30 nJ per pulse at repetition rates up to 250 kHz. This energy may be increased to over 50 nJ per pulse with the addition of an RF amplifier to increase the dumping efficiency of the AOM. We do not require large pulse energies initially, but will implement the upgrade as needed. The pulses exhibited a spectral bandwidth of 65 nm FWHM, and had an overall wavelength spread of over 100 nm.

Our original intent was to copy the design of Wiersma [9], but we found that their design yielded a very poor contrast ratio of 20-30:1 between the intensity of the dumped pulse and the pulses immediately before and after the dumped pulse. This would have led to distortion of any time dependent signals measured using the dumped pulses in a pump/probe apparatus. The problem was traced back to the resonator design requirements imposed by the Kerr lens modelocking which occurs in TiS lasers. These requirements result in a relatively large beam waist in the AOM, thereby degrading the ability of the AOM to cleanly dump a single pulse from the pulse train. Extensive numerical simulations of alternative laser resonator configurations revealed a configuration which would yield a smaller waist in the AOM while still allowing Kerr lens modelocking. After implementing the new resonator design, contrast ratios of 200:1 have been achieved.

The very wide bandwidth pulses produced by the oscillator are split into a pump pulse (95%) and a local oscillator pulse (5%). Each of these pulses in then directed into independent spectral filters, each of which comprises a pair of SF11 prisms and a variable width translatable slit. After the second prism the 100 nm laser pulse spectrum is spatially dispersed to a width of about 12 mm. By setting the slit to about 1 mm, we can select 12-15 nm FWHM sections of the spectrum for use as the pump and local oscillator pulses. Autocorrelation measurements have yielded autocorrelation widths of about 130 fsec FWHM. The filtering mechanism leads to spectral profiles which are roughly trapezoidal, and Fourier transforming the spectra has shown that the filtered pulses are about 100 fsec FWHM and quite close to transform limited (within experimental uncertainty).

In summary, we can produce 100 fsec pulses with 12-15 nm bandwidth over the range of 750 nm to 860 nm. More importantly, the pump and local oscillator pulses are independently tunable, which is essential for implementation of the Chronocyclic Spectrometer.

DESCRIPTION OF Ti:S LASER REGENERATIVE AMPLIFIER

For situations where higher pulse energies are required, we have designed, and are completing the process of constructing, a cw-pumped acoustooptically Q-switched TiS regenerative amplifier. In this configuration the oscillator described above is operated with an output coupler instead of a cavity dumper. A second TiS resonator is pumped by the same argon ion laser which pumps the oscillator. This second resonator contains an acoustooptic Q-switch as well as a Bragg cell. The Q-switch prevents lasing in the amplifier cavity until it is switched off and a seed pulse from the oscillator is injected into the amplifier by the Bragg cell. Prior to injection this pulse is spectrally filtered and temporally stretched by a grating pulse stretcher. The spectral filtering allows tuning of the regenerative amplifier output, while the temporal stretching is required in order to avoid nonlinear effects in the amplifier by reducing peak intensities. This stretched pulse makes 20-25 round trips through the amplifier (until the amplifier gain is saturated) and is then switched out, having been amplified to several uJ. At this point the Q-switch is turned on to prevent lasing until the next seed pulse is injected. The amplified pulse will be recompressed by a grating pulse compressor to a transform limited pulse duration of ca. 100 fsec. The resulting output will be 1-2 uJ at repetition rates from several kHz up to 250 kHz tunable from 770 nm to 840 nm.

PINHOLE MASK FABRICATION

Spatial resolution in the Chronocyclic Spectrometer is to be provided by pinhole overcoating, in which a sample is covered with a metal mask that is permeated with 50 nm-radius apertures which define the region of optical excitation or signal collection. The pinhole apertures are fabricated by spincoating the surface of a sample with a dilute suspension of monodisperse polystyrene spheres. A metal film of thickness less than the sphere radius is then deposited by vacuum evaporation. The latex spheres are subsequently removed by sonication resulting in a near field mask with a random spatial distribution of apertures. A scanning electron micrograph of 1.0 micrometer and

0.5 micrometer pinholes that we have produced using this technique is shown in Figure 3.

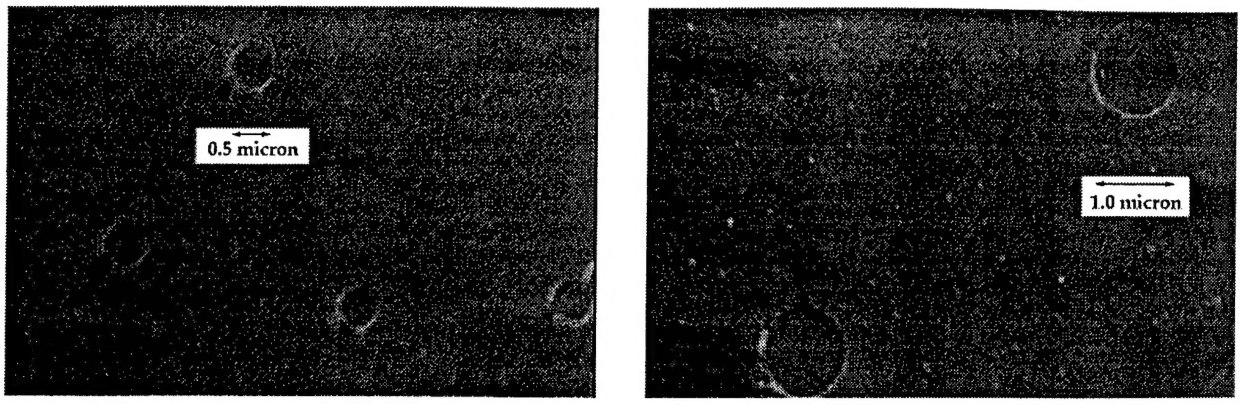

Figure 3: Scanning electron micrograph of pinhole masks

MANUSCRIPTS SUBMITTED DURING REPORTING PERIOD:

1. David. S. Alavi, M.G. Raymer, and Peter C. Sercel, "Two color time resolved pump/probe spectroscopy using a cavity dumped Kerr lens modelocked Ti:sapphire oscillator", in preparation.

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See attached DD Form 882.

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